

Lateral Grain Growth of Poly-Si Films with a Specific Orientation by an Excimer Laser Annealing Method

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(Received July 21, 1993; accepted for publication September 18, 1993)

Dramatic lateral grain growth of nondoped poly-Si films (maximum grain size: $\sim 4.5 \mu\text{m}$, film thickness: 500 Å) with strong crystallographic (111) orientation on glass substrates has been achieved using an excimer laser annealing method, namely at low temperature below 400°C and in a processing time shorter than a second, for the first time. The surface morphology of these poly-Si films was very smooth and the crystallinity was excellent with minimal internal defects. These poly-Si films have monomodally distributed grain sizes, with an average grain size of around 1.5 μm . As a result of experimental study, we speculate that the basic driving force behind this lateral grain growth was surface free energy anisotropy, as the same mechanism was observed in high-temperature furnace annealing of doped poly-Si thin films.

KEYWORDS: poly-Si, excimer laser annealing, lateral grain growth, grain size, crystallographic orientation, surface free energy, anisotropy

1. Introduction

Development of low-temperature processed ($< 600^\circ\text{C}$) polycrystalline silicon (poly-Si) thin-film transistors (TFT) is expected as a solution to realizing future giant-microelectronic devices such as large-area, high-definition liquid crystal displays.¹⁾ In particular, low-temperature poly-Si fabrication technique capable of utilizing a relatively inexpensive substrate such as glass is thought to be fundamental to the development of this technology.

Among the various approaches being examined, excimer laser annealing is being viewed with particular hope as a means of realizing a high-performance poly-Si TFT on glass substrate for giant-microelectronics.²⁻⁷⁾ Because this method is of extremely short duration ($< 100 \text{ ns}$) and is a shallow melt-regrowth process, it can prevent the introduction of thermal damage to the glass substrate^{3,8)} and there also is no need to worry about the thermal compaction problem, which is a serious issue in the solid phase crystallization method. Moreover, due to the high power and large beam size of the excimer laser, this method offers the basic potential to be a high-throughput process. However, one serious problem is that poly-Si films obtained by this technique have a small grain size of less than 1000 Å.⁹⁾ This is because the solidification velocity during excimer laser annealing is extremely high.⁹⁾

To solve the grain size problem, we previously proposed a new concept in the excimer laser annealing method—artificial control of the solidification velocity—to enlarge the grain size and to enhance the field effect mobility, for the first time.³⁾ This method is based on newly developed three-dimensional thermal analysis and experimental results. We were able to specifically clarify two parameters effective in reducing the solidification velocity of molten Si without adopting a special substrate structure. One was low-temperature ($\leq 400^\circ\text{C}$) substrate heating during excimer laser irradiation and the other was thinning of the a-Si thickness

($\sim 500 \text{ \AA}$) as a starting material.

Using this method and controlling the above parameters, we have obtained a large grain size of over 5000 Å and a high field-effect mobility of 280 $\text{cm}^2/\text{V}\cdot\text{s}$.¹⁰⁾ We also clarified that this method effectively improves the uniformity of the laser-irradiation overlap region, the most serious problem when applying excimer laser recrystallized poly-Si films to giant-microelectronics.⁴⁾

Recent studies¹¹⁾ demonstrate that large-grain-size poly-Si film ($\sim 2 \mu\text{m}$) can be obtained by means of excimer laser irradiation not only from the front side but also from the back side to reduce the solidification velocity of molten Si. In principle, this technique involves the same concept as that which we proposed.³⁾ As an ultimate method for reducing the vertical solidification velocity during excimer laser annealing, Choi *et al.* reported extremely large-grain poly-Si films ($\sim 50 \mu\text{m}$) using superthin Si film formed on the thin SiO_2 membrane as a substrate.¹²⁾

However, the two methods described above seem to be particularly difficult to apply to actual factory processing because a special apparatus or special substrate structure is needed. Including the above two methods, excimer laser recrystallized poly-Si films have not been realized with orientation controlled as before.

In this paper, we report dramatic lateral grain growth of poly-Si films on glass substrates with both orientation and grain size controlled, for the first time. These films were produced using our proposed new method combined with multishot laser annealing. We also discuss and speculate upon the mechanism of this lateral grain growth phenomenon based on extensive experimental study and on the analogy of a similar phenomena observed in other methods.

2. Experimental

Nondoped amorphous silicon (a-Si) film (500 Å) was deposited on a quartz substrate by the plasma-enhanced chemical vapor deposition (PECVD) method.

The deposition was performed using 100% SiH₄ decomposition at a substrate temperature of 170°C, an RF power density of 8 mW/cm², a pressure of 26.6 Pa and a gas flow rate of 22 sccm. Prior to laser irradiation, the a-Si was dehydrogenated by annealing in a vacuum chamber at 450 to 550°C. The a-Si was recrystallized by ArF excimer laser pulses ($\lambda=193$ nm, $\tau=17$ ns) in a vacuum chamber ($<10^{-4}$ Pa). A 3 × 3 mm laser beam was unified by beam-homogenized optics. The uniformity of laser beam intensity was set within $\pm 5\%$.

The crystallinity, microstructure and orientation of the poly-Si films were evaluated by scanning electron microscopy (SEM), transmission electron microscopy (TEM), transmission electron diffraction (TED) and by their X-ray diffraction pattern.

3. Results

Figure 1 shows SEM images of Secco-etched poly-Si films recrystallized using varying numbers of shots of excimer laser to anneal nondoped 500-Å-thick a-Si, where the substrate temperature was 400°C and the laser energy density was 350 mJ/cm².

In the case of small numbers of laser shots, such as two or eight shots, the grain size was enlarged to about 5000 Å due to the reduction in the solidification velocity of the molten Si, as reported previously.¹⁰ However, when the number of laser shots was increased further, it was found that dramatic lateral grain growth occurred, as this figure illustrates. The poly-Si film annealed using 64 shots exhibited a maximum grain size of about 3.0 μm and we note that the largest grains had a length of about 4.5 μm in the case of 128 shots. This value is about ten times that for films we reported previously.¹⁰ Similar large grain sizes in poly-Si films were reported using solid phase crystallization of a-Si in a furnace annealing at ~ 600 °C.¹³⁻¹⁶ But the grains obtained in this case were of dendritic structure and have many types of internal defects, such as twin, dislocation and stacking faults. In contrast, the poly-Si films we obtained showed very few defects within the bulk of the grain, as shown in these SEM images, and exhibited excellent crystallinity. TEM micrographs obtained from these poly-Si films also confirmed

low defect densities within grains, as the same result reported previously.¹⁰

Figure 2 shows the transition of grain size distribution with the above lateral grain growth as revealed in Secco-etched SEM images. Using an image analysis processor, the area of each grain was calculated and individual grain size was taken to be the diameter of an area-equivalent circle. The number of grains was at least 500. As shown in this figure, poly-Si films annealed using two shots exhibited almost lognormally distributed grains with an average grain size of about 2000 Å. The distribution of 64-shot-annealed poly-Si films, however, shifted to bimodal. This distribution seemed to occur during the process of lateral grain growth. Finally, 128-shot-annealed poly-Si films again

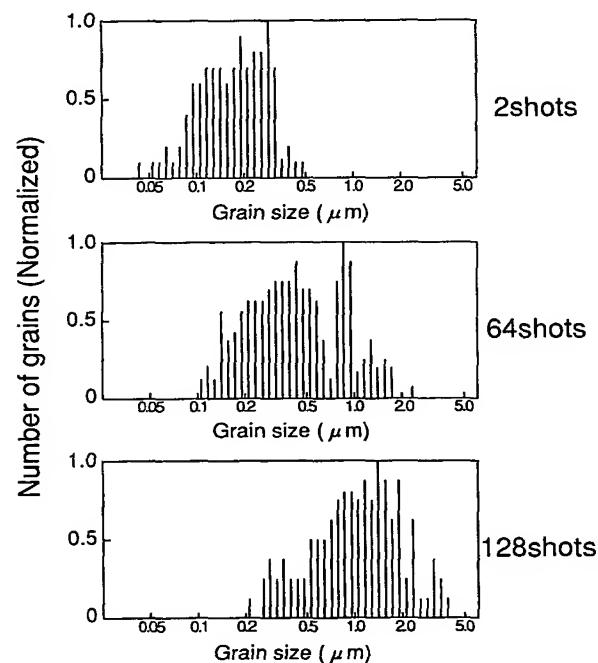


Fig. 2. The transition of grain size distribution using various numbers of shots to perform excimer laser annealing (laser energy density: 350 mJ/cm², substrate temperature: 400°C).

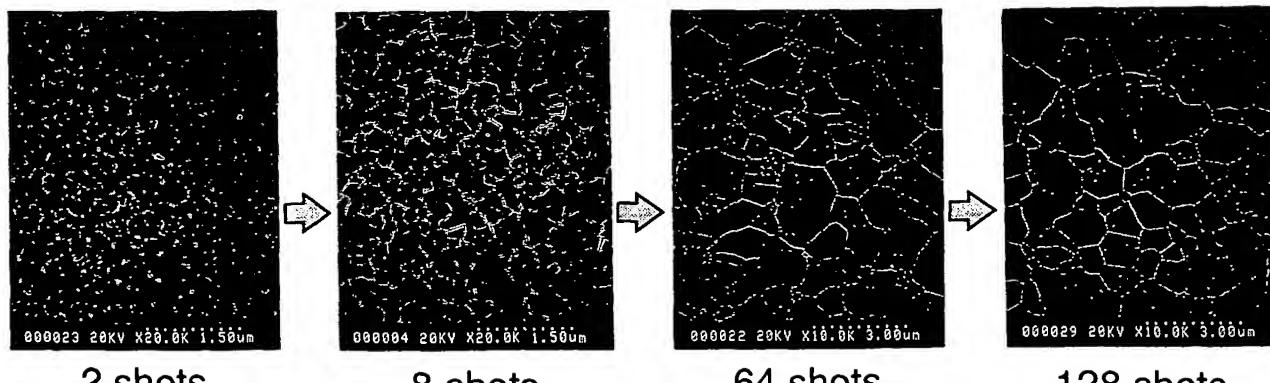


Fig. 1. SEM images of Secco-etched poly-Si films recrystallized using various numbers of shots to perform excimer laser annealing (laser energy density: 350 mJ/cm², substrate temperature: 400°C).

exhibited almost monomodal grain size distribution with an average grain size of about $1.5 \mu\text{m}$.

This result clearly shows that this method offers an additional advantage in terms of the distribution of grain size compared with the solid phase crystallization method. In thermally crystallized poly-Si films, it is reported that grain size was widely distributed and that the frequency decreased monotonically as the grain size became larger.^{17,18)} Thus, device characteristics were reported to be inhomogeneous in thin-film transistors fabricated using such poly-Si films, when the channel length becomes closer to the grain size.¹⁹⁾ On the contrary, using the poly-Si films we obtained, device characteristics are expected to become homogeneous due to the shrunken grain size distribution and controlled orientation.

Figure 3 shows cross-sectional SEM images of large-grain poly-Si films obtained by 64-shot annealing. As shown in this figure, the grains are entirely of a large columnar structure that is many times larger than the film thickness. The surface morphology of these films is also quite smooth rendering them well-suited to thin-film transistor applications.

To investigate the mechanism of this lateral grain growth phenomenon, we evaluated these poly-Si films by X-ray diffraction spectra. As shown in Figure 4, both two-shot and eight-shot laser-annealed poly-Si films appeared to have random polycrystalline orientation, as reported earlier.¹⁰⁾ But as the number of laser shots was increased, only (111) peak intensity radically increased, and the (110) peak simultaneously disappeared completely. This result indicates that (111) crystallographic orientation occurred with lateral grain growth. We also confirmed that the selected-area transmission electron diffraction (TED) of these laterally grown grains indicated a (111) crystallographic orientation.

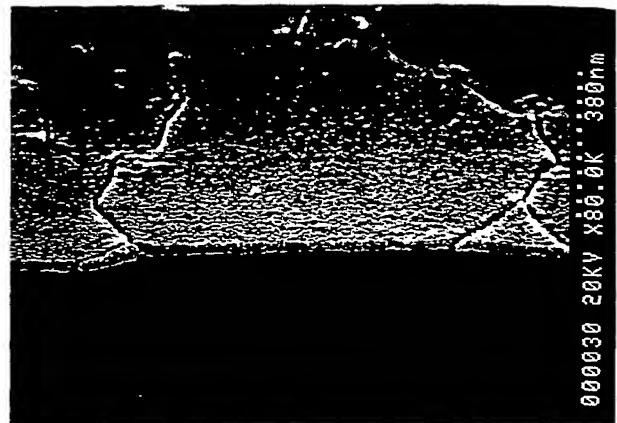


Fig. 3. Cross-sectional SEM images of Secco-etched poly-Si films recrystallized by 64-shot excimer laser annealing (laser energy density: 350 mJ/cm^2 , substrate temperature: 400°C).

Figure 5 shows SEM images of Secco-etched poly-Si films recrystallized at different substrate temperatures ($RT \sim 400^\circ\text{C}$) during excimer laser annealing of non-doped 500-Å-thick a-Si, where the number of laser shots was 128. As shown in this figure, the lateral grain growth phenomenon was observed at substrate temperatures over 200°C and the grain size was noted to increase with the substrate temperature.

Figure 6 shows the X-ray diffraction pattern for these films. In corresponding SEM images, (111) crystallographic orientation was shown to occur over a substrate temperature of 200°C . These results indicate that this lateral grain growth phenomenon occurs over a relatively wide range of laser annealing conditions and has temperature dependence.

We also attempted using 1000-Å-thick a-Si film as a starting material. In this case, only a slight enhance-

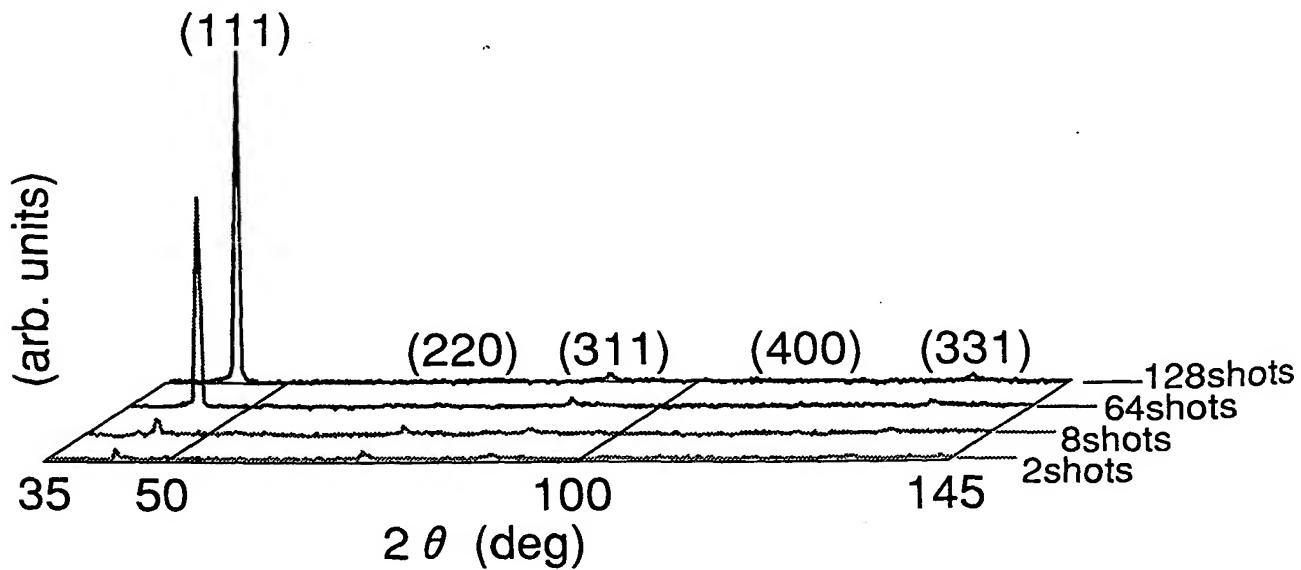


Fig. 4. X-ray diffraction spectra of poly-Si films recrystallized using various numbers of shots to perform excimer laser annealing (laser energy density: 350 mJ/cm^2 , substrate temperature: 400°C).

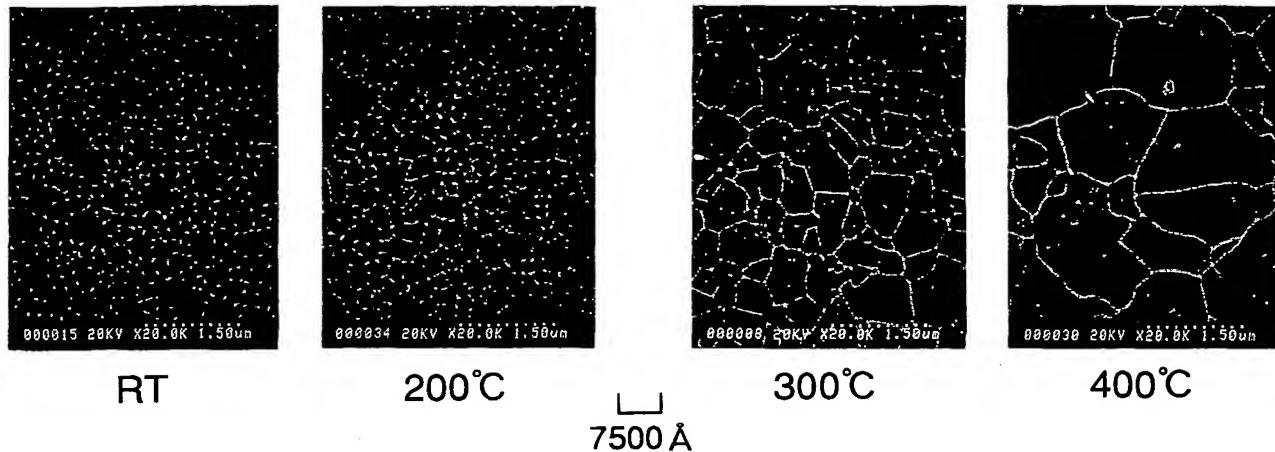


Fig. 5. SEM images of Secco-etched poly-Si films recrystallized created at different substrate temperatures (laser energy density: 350 mJ/cm^2 , numbers of laser shots: 128).

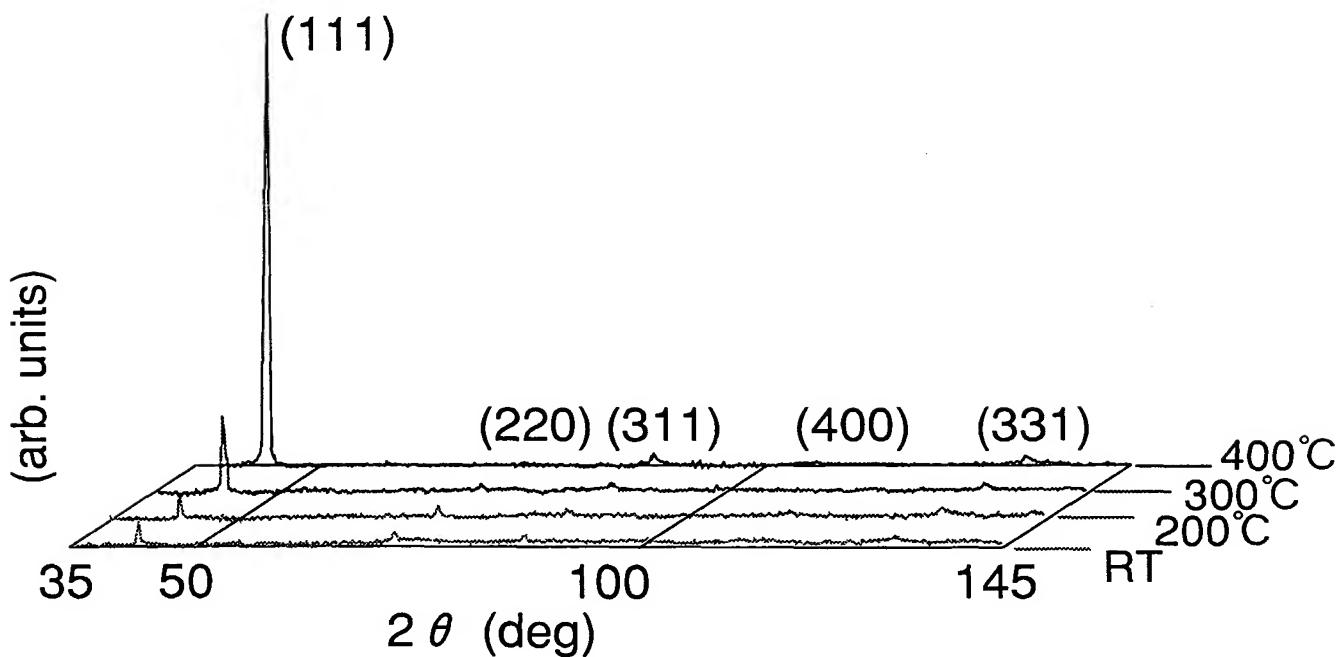


Fig. 6. X-ray diffraction spectra of poly-Si films recrystallized at different substrate temperatures (laser energy density: 350 mJ/cm^2 , numbers of laser shots: 128).

ment of grain size was observed and notable (111) crystallographic orientation was not observed. These results indicate that film thickness is also an important parameter in realizing dramatic lateral grain growth during excimer laser annealing.

4. Discussion

The above experimental results clearly show that this lateral grain growth phenomenon was caused by a certain anisotropy which exhibited a (111) preferential orientation because it is known that normal grain growth saturates in grain size grow to be approximately equal to the film thickness. There are two possible explanations for this anisotropy-oriented grain growth.

One is the anisotropy of surface free energy. The other is the anisotropy of the melting temperature, which is different from the crystallographic orientation in Si. As regards the anisotropy of the melting temperature, it is known that (100) textured crystalline structures have a slightly higher melting temperature compared to other textures and can coexist with molten Si.²⁰⁾ It has been asserted that this small difference in melting temperature might lead to the (100) orientation observed in Si graphoepitaxy by CW-laser and strip heater recrystallization.²⁰⁾ But it has also been reported that over a narrow range of laser power, the silicon appeared to be partially molten and only under these conditions was the preferential orientation observed. This liquid-solid

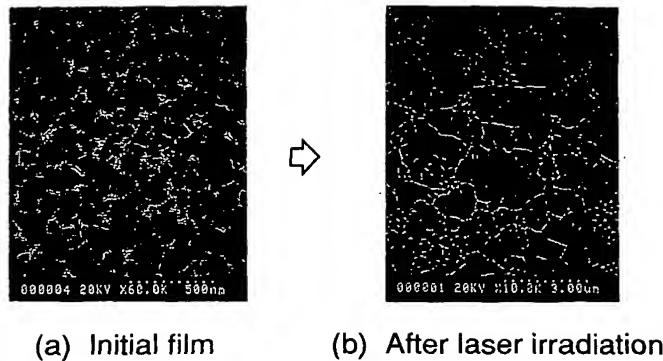
coexistence is stable only when the Si contacts oxide on both sides.

But the poly-Si films we obtained have a (111) crystallographic orientation, as described above, and were formed under a relatively wide range of conditions. Consequently, it is difficult to explain our (111) preferential orientation poly-Si films as a product of the anisotropy of the melting temperature.

The second possibility, that is, the anisotropy of surface free energy, is a phenomenon quite similar to that in the nondoped poly-Si films we obtained. Referred to as secondary grain growth in thin films, this phenomenon has also been observed in the high-temperature ($\sim 1200^{\circ}\text{C}$) and long-time (~ 20 min) thermal annealing of thin (< 1000 Å) doped poly-Si films reported by C. V. Thompson and colleagues.²¹⁻²⁴ According to their experimental results and developed theory, grain growth in thin films is affected by anisotropy of the surface free energy, and grains that exhibit secondary grain growth will have orientations which minimize surface free energy.²² It is known that (100) texture minimizes the energy of the Si/SiO₂ interface and that a (111) texture minimizes the energy of the Si free surface.²⁵ It is also thought that the magnitude of the free surface energy is larger than the magnitude of the Si/SiO₂ interface energy.²¹ The results of doped thin poly-Si film lead to: (1) grains much larger than the film thickness (average grain size: ~ 2 μm) with monomodally distributed size, (2) grains with uniform crystallographic (111) orientation, and (3) low densities of defects including dislocations and twins. It has also been reported that the rate of secondary grain growth should increase with: ① decreasing film thickness, ② increasing temperature, ③ increasing average surface energy anisotropy, and ④ increasing average grain boundary mobility (e.g., doping with phosphorus or arsenic).²³

Considering the similarity of characteristics and conditions between the above lateral grain growth and the poly-Si films we obtained, we speculate that the basic driving force behind the lateral grain growth observed was also surface free energy anisotropy.

To confirm this assumption, we examined two types of starting materials other than a-Si. One was an as-deposited thin poly-Si film created by the LPCVD method at 620°C . This film showed a nearly columnar fiberlike grain structure in which the average grain size in the plane of the film is smaller than the average grain size perpendicular to the plane of the film. The other starting material was a completely cylindrical grain structure in which the average grain diameters were roughly equal to or several times larger than the film thickness, as shown in Fig. 7(a). This material was fabricated by excimer laser annealing of a-Si (500 Å thick) films at room temperature, where the laser energy density was 300 mJ/cm². This film had an average grain size of around ~ 1000 Å. Only in the latter case was a dramatic lateral grain growth phenomenon (~ 3 μm) observed. Conditions were irradiation using multishot (64 shots) excimer laser annealing with a substrate temperature of 400°C , as shown in Fig. 7(b).



(a) Initial film (b) After laser irradiation

Fig. 7. SEM images of Secco-etched poly-Si films. (a) Initial film: excimer laser annealing of a-Si (500 Å) at room temperature where laser energy density was 300 mJ/cm². (b) After laser irradiation of film (laser energy density: 350 mJ/cm², substrate temperature: 400°C , numbers of laser shots: 64).

This result also supports the importance of initial grain structure, in other words, surface free energy, in, the lateral grain growth phenomenon we observed.

However, there are two clearly different characteristics compared with the earlier report. First, the secondary grain growth in furnace annealing has been observed only in films doped with donor impurities such as phosphorus and arsenic.²³ On the contrary, the poly-Si films we observed were nondoped. Second, secondary grain growth in furnace annealing has been conducted using isothermal annealing for at least 20 min.²⁴ On the other hand, the lateral grain growth phenomenon we observed occurs over extremely short periods (< a second). In the case of high-temperature annealing, samples doped to below 3×10^{20} cm⁻³ did not exhibit secondary grain growth even at temperatures as high as 1350°C .²³ The reason for the necessity of dopant for secondary grain growth is explained in terms of an increased atomic mobility of grain boundaries due to changes in point defect concentrations resulting from changes in the Fermi level.²⁴ Grain boundary mobility was found to correspond roughly scales with self-diffusivity. It has been suggested that phosphorus diffusion is the rate-limiting step in secondary grain growth in silicon. Thus, a process of long duration (> 20 min) is required.

In our previous study, three-dimensional thermal analysis showed that the region of grain boundary melted completely and that the grain region was pinned with the Si melting temperature (not fully molten) in the case of excimer laser irradiation to poly-Si film where the substrate temperature was 400°C .⁴ From this result, and based on the phenomenon observed in the current study, we conclude that it is possible to explain that this complete melting of the grain boundary enhances the atomic mobility of the grain boundary without doping, as well as the speed of the lateral grain growth phenomenon.

5. Conclusions

We have succeeded in obtaining excellent-crystallinity large-grain thin (500 Å) nondoped poly-Si films with

both orientation and grain size controlled on glass substrates, for the first time, using the proposed new excimer laser annealing method combined with multishot laser annealing, namely a low-temperature and short-duration process. The films produced had a maximum grain size of $4.5 \mu\text{m}$ with a strong (111) crystallographic orientation and a monomodally distributed grain size of $1.5 \mu\text{m}$, on average. The surface morphology of these films was very smooth and the crystallinity was excellent with minimal internal defects.

As a result of experimental study and on the analogy of a similar phenomenon observed in high-temperature furnace annealing of doped poly-Si thin films, we speculate that the basic driving force behind this lateral grain growth was surface free energy anisotropy, and that complete melting of the grain boundary enhances the grain boundary mobility without doping, as well as the speed of the lateral grain growth phenomenon.

Our achievement will be promising as a technique for fabricating high-speed drive circuits on glass substrates.

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